Effect of impurities on supersolid condensate: a Ginzburg-Landau approach

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Basing our arguments on a wave function that contains both positional and superfluid order, we propose a Ginzburg-Landau functional for a supersolid with the two order parameters necessary to describe such a phase: density $n_B(\mathbf{r})$ and supersolid order parameter ψ_V. We argue that adding lighter ^{3}He atoms to a ^{4}He supersolid produces attractive regions for vacancies, leading to patches of higher T_c. On the other hand, the supersolid stiffness decreases in this granular state with increased ^{3}He disorder. Both effects are linear in ^{3}He concentration.

INTRODUCTION

Recent experiments by Chan and Kim [1, 2] have generated renewed interest in the possibility of the existence of a supersolid phase in ^{4}He. There is pioneering theoretical work by, among others, Andreev and Lifshitz [3], Reatto [4], Chester [5], Leggett [6] and Anderson [7]. Recently, Anderson [8] and Anderson, Brinkman and Huse [9] have discussed the problem. Recent developments have been reported at a KITP workshop and are available online [10]

The main purpose of purpose of this note is to develop an approach that allows a discusssion of the effect of ^{3}He impurities on a ^{4}He supersolid. In what follows, we show how the approach leads to a Ginzburg-Landau functional for the supersolid. We propose to follow the collective degrees of freedom that are candidates for the supersolid properties, namely the vacancies that are intrinsic to an incommensurate [9] solid. Thus, we follow Andreev and Lifshits [3], and more recently Anderson [8], who argued that the supersolid requires some defects, which are inherently present in the phase at $T = 0$. Vacancies and interstitials are obvious candidate defects in the ^{4}He solid. For simplicity, we focus here on vacancies [11]. In this picture, the lattice sites are equivalent, there is no disorder and yet the He solid is not commensurate.

Thus, we can consider the ground state of ^{4}He at $T = 0$ as a state in which there is a non-zero concentration of vacancies. Although a localized vacancy may require a non-zero activation energy, the fact that they are mobile (the more so due to the large zero-point motion of the He atoms) can bring the bottom of their band to zero energy. For any non-zero concentration n_V of vacancies, they will form a Bose condensate at $T = 0$. However, to properly describe the quantum nature of the condensate we have to address not the densities but the quantum amplitudes of the relevant degreees of freedom. Thus the Ginzburg-Landau (GL) functional for the supersolid deals with the vacancy Bose field $\psi_V = \sqrt{n_V}\exp(i\alpha)$ as an order parameter.

WAVE FUNCTION FOR A SUPERSOLID

In this section, we discuss wave functions that display solid order with, at the same time, a Bose condensed fraction of vacancies.

We start with a lattice state of hard-core bosons; the boson occupancy cannot exceed one per lattice site. The operator b_i^+ creates a ^{4}He atom at site i of the lattice. The commensurate crystal wave function is then

$$|\Psi_0\rangle = \prod_i b_i^+|0\rangle = |\uparrow,\uparrow,...\uparrow\rangle, \tag{1}$$

corresponding to a "ferromagnetic" state, with no vacancies. For some purposes, it is convenient to use a spin-1/2 representation [12]:

$$S_i^+ = b_i^+,\ S_i^- = b_i,\ S_i^z = b_i^+ b_i - 1/2. \tag{2}$$

To introduce vacancies, a simple procedure would be to apply $\sum_i S_i^-\ N_V$ times to create N_V vacancies. However, it has been understood since BCS and was pointed out in the present context by Anderson [8] that a phase coherent superfluid state must be a linear combination of states of different particle number. To achieve this, we apply a tilt operator to the "ferromagnetic" commensurate state. The rotation operator in the spin representation is (now using Pauli operators)

$$\prod_i \exp(i\boldsymbol{\sigma}_i\cdot\hat{\mathbf{a}}_i\phi_i/2) = \prod_i[\cos\frac{1}{2}\phi_i + i(\boldsymbol{\sigma}_i\cdot\hat{\mathbf{a}}_i)\sin\frac{1}{2}\phi_i]. \tag{3}$$

Here, ϕ_i is the tilt angle at the i-th site and $\hat{\mathbf{a}}_i$ is the rotation axis, which lies in the x, y plane. The latter's angle with respect to the y axis will be denoted by α_i. Since $\hat{\mathbf{a}}_i \perp \hat{\mathbf{z}}$,

$$\boldsymbol{\sigma}_i\cdot\hat{\mathbf{a}}_i = (S_i^+ a_i^- + S_i^- a_i^+). \tag{4}$$

With $a_i^\pm = a_x \pm i a_y = \pm i\exp(\pm i\alpha_i)$, we get

$$\boldsymbol{\sigma}_i\cdot\hat{\mathbf{a}}_i = ie^{i\alpha_i}b_i - ie^{-i\alpha_i}b_i^+. \tag{5}$$

We operate with Eq. (3) on $|\Psi_0\rangle = \prod_i b_i^+|0\rangle$ and use Eq. (5) to find the tilted state

$$\begin{aligned} |\Psi\rangle &= \prod_i [\cos\frac{1}{2}\phi_i + i(ie^{i\alpha_i})\sin\frac{1}{2}\phi_i\ b_i]b_i^+|0\rangle \\ &= \prod_i [\cos\frac{1}{2}\phi_i\ b_i^+ - e^{i\alpha_i}\sin\frac{1}{2}\phi_i]|0\rangle, \end{aligned} \tag{6}$$

where we have used the fact that because of the hard-core constraint, $S_i^+ = b_i^+$ gives zero acting on $|\Psi_0\rangle$. The state $|\Psi\rangle$ is a linear combination of states with specified phases and different numbers of vacancies; it is normalized and the expectation value of vacancy occupation is:

$$n_V^i = 1 - \langle b_i^+ b_i\rangle = \sin^2\frac{1}{2}\phi_i \tag{7}$$

The vacancies are mobile due to rearrangements of the atoms, which is facilitated by their large zero-point motion. We model the kinetic energy of vacancies with a simple nearest-neighbor hopping hamiltonian [13]. Its expectation value in $|\Psi\rangle$ is [14]:

$$\begin{aligned} KE &= -t\sum_{\langle ij\rangle}\langle b_i^+ b_j + h.c.\rangle \\ &= (-t/2)\sum_{\langle ij\rangle}\sin\phi_i\sin\phi_j\cos(\alpha_i - \alpha_j). \end{aligned} \tag{8}$$

With all ϕ_i, α_i the same, $|\Psi\rangle$ is a state with both solid order and a phase coherent vacancy contribution. The vacancy order parameter is

$$\psi_V = \langle b_i\rangle = \frac{1}{2}\sin\phi e^{-i\alpha}. \tag{9}$$

In the spin language, when all ϕ_i, α_i are the same, $|\Psi\rangle$ is a state with maximal total spin $S = M/2$, where M is the number of lattice sites. The expectation value of $S^z = \sum S_i^z$ is $\langle S^z\rangle = (M/2)\cos\phi$.

GINZBURG-LANDAU FUNCTIONAL

We start with with the postulate that the GL functional of a supersolid phase in ^{4}He has to contain *two* order parameters: density $n_B(\mathbf{r})$ and superfluid amplitude $\psi_V(\mathbf{r})$. Although there are only ^{4}He atoms, these atoms particpate in two distinct phenomena: solid and superfluid. In the region of the experimental phase diagram [1], where one enters a possible supersolid phase from the solid at $T \sim 100mK$ at fixed pressure, solid order is presumably already well established and hence is well outside of the GL regime. Thus, we concern ourselves with a possible second-order transition across a normal solid to supersolid transition line. The amplitude $|\psi_V(\mathbf{r})|$ is small at the transition and is in the GL regime. The wave function $|\Psi\rangle$ in Eq. (6) offers a unified description of both solid order with periodically modulated density $n_B(\mathbf{r})$ and vacancies with the order parameter ψ_V plus the constraint $n_B(\mathbf{r}) + n_V(\mathbf{r}) = 1$. In the discussion below we focus on the ψ_V field.

Toward developing the GL approach, we assume that the vacancy density is small (as is consistent with the experimental situation) so that the tilt angle ϕ is small. To estimate the superfluid stiffness, we examine the KE as the phase α acquires a slow variation from site to site. From Eqs. (8,10)and small ϕ, we get

$$\Delta\, KE = (t/a_0)\int d\mathbf{r}|\psi_V|^2(\nabla\alpha)^2, \tag{10}$$

where a_0 is the lattice spacing in a simple cubic lattice. We see that the superfluid stiffness is determined by the hopping amplitude t and the density of vacancies $|\psi_V|^2$.

Next we examine the effect of a potential that couples to the ^{4}He density, $H_1 = U_i b_i^+ b_i$. From Eq. (7),

$$\begin{aligned} \langle H_1\rangle &= \sum_i U(i)\cos^2\frac{1}{2}\phi_i \\ &\approx -(1/\Omega)\int d\mathbf{r}\, u(\mathbf{r})\sin^2\frac{1}{2}\phi(\mathbf{r}) \\ &\approx -(1/\Omega)\int d\mathbf{r}\, u(\mathbf{r})|\psi_V(\mathbf{r})|^2, \end{aligned} \tag{11}$$

where Ω is the unit cell volume, $u(\mathbf{r}) = \Omega\sum_i U_i\delta(\mathbf{r} - \mathbf{r}_i)$ is the potential energy and we ignored the constant $\int d\mathbf{r}\, u(\mathbf{r})$. In the last line, we took advantage of the approximation that ϕ is small so that, from Eq. (10), $|\psi_V|^2 \approx n_V = \sin^2\frac{1}{2}\phi$. In our proposed state $|\Psi\rangle$, a potential that is *repulsive* for ^{4}He atoms would be *attractive* for vacancies, tending to increase their local density.

Our discussion leads to the total GL free energy density

$$\begin{aligned} F(\mathbf{r}) &= [a(\frac{T}{T_c^0} - 1) - u(\mathbf{r})/\Omega]|\psi_V(\mathbf{r})|^2 + (t/a_0)|\psi_V(\mathbf{r})|^2|\nabla\alpha|^2 \\ &+ b|\psi_V(\mathbf{r})|^4. \end{aligned} \tag{12}$$

The quartic term in F represents the restriction against putting two vacancies in the same place, as well as dynamical interaction terms.

EFFECT OF DISORDER

Chan et al [2] find the remarkable result that by adding ^{3}He atoms in the ^{4}He solid, the measured T_c grows and the superfluid stiffness ρ_s drops as a function of ^{3}He concentration. The data is reproduced here in Fig. 1.

Let us therefore address the effect of disorder on the supersolid within the GL framework. In particular, because of its experimental relevance, we are interested in the effect of ^{3}He impurities on the superfluid properties. One might expect that disorder would suppress the supersolid state as it would interfere with the phase ordering as it

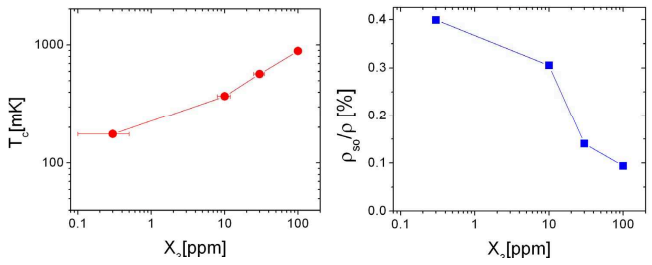

FIG. 1: Dependence of transition temperature and superfluid stiffness on ^{3}He concentration. Data of Chan et. al. [2], also quoted in [1]

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does in superconductors. However if the supersolid arises due to the presence of vacancies, then the presence of defects that *create* vacancies or that can increase the local concentration of vacancies could lead to enhanced superfluidity. Here, we offer some speculations on the effects of a non-zero concentration of ^{3}He atoms on the transition temperature and superfluid stiffness.

1) The presence of lighter ^{3}He atom defects increases the zero point fluctuations locally; this repels nearby host atoms. As we have seen, a repulsion for ^{4}He atoms is an attraction for vacancies; this increases the local vacancy density as seen in Fig. 2. This leads to an increase in T_c. To first approximation, the effect would be linear in the ^{3}He concentration. 2) At the same time, there would be an effect on the superfluid stiffness ρ_s. We suggest that ρ_s would decrease with increased ^{3}He concentration because ^{3}He sites would produce attractive sites for the superfluid condensate and localize it more. While ^{3}He creates puddles of enhanced supersolid condensate, the stiffness as a reflection of the global phase rigidity would drop on average with increased ^{3}He doping. In other words, to the extent that substitutional impurities inhibit the exchanges of ^{4}He atoms that are required to

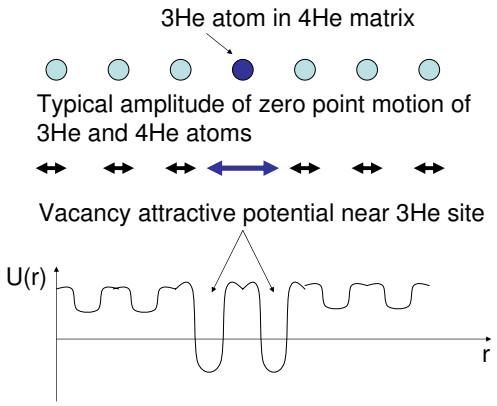

FIG. 2: When included in the ^{4}He lattice, ^{3}He atoms will push the host atoms away. This redistribution is equivalent to an attractive potential for vacancies, thus generating a larger vacancy density near ^{3}He.

enable vacancy hopping, we expect that any foreign atom substitution will diminish the effective t, leading to a reduction in superfluid stiffness. Again, the effect will be linear in impurity concentration, see Fig(1).

So we propose that a ^{3}He site is attractive for vacancies and we model the effect of ^{3}He substitution as a random attractive potential for vacancy density with the potential $u(\mathbf{r})/\Omega = +\lambda\rho_3(\mathbf{r})$, where $\lambda\,(>0)$ is the coupling strength of the effective ^{3}He – ^{4}He repulsion. The corresponding contribution to the GL free energy density is

$$F_3 = -\lambda n_3 |\psi_V(\mathbf{r})|^2. \tag{13}$$

Here, we have taken an annealed average, $\langle\rho_3(\mathbf{r})\rangle = n_3$ as the average density of ^{3}He atoms.

We see, from Eqs. (12-14), that the effect of ^{3}He substitution is to increase the transition temperature from T_c^0 to

$$T_c = (1+\frac{\lambda}{a}n_3)T_c^0, \quad \delta T_c/T_c^0 = \frac{\lambda\,\delta n_3}{a} \tag{14}$$

an effect that is linear in ^{3}He concentration n_3. One could estimate the parameters by examining the experimental data. In fact the numbers from Chan et al [2] show an enormous effect. From the data, Fig. 2, we find that shift $\delta T_c = 150$ mK, or $\delta T_c/T_c \approx 0.75$ for an increase in ^{3}He concentration δn_3 of about 10 ppm. Put differently, the as defined critical temperature nearly doubled upon adding 0.001% impurities. To date, no known superfluid states exhibit such a stong sensitivity to impurities. This is one of the many puzzles in this system [15].

Now we turn to stiffness corrections. We argued above that the stiffness term will soften in the presence of ^{3}He defects. The effective hopping t in the GL free energy density of Eq. (13) is then $t = t_0(1-g\,n_3/n_V)$ with $g>0$. Since $\rho_s \propto t|\psi_V|^2$, we conclude that

$$\rho_s = \rho_s^0(1-g\,n_3/n_V), \quad \rho_s^0 = 2(t_0/a_0)\;|\psi_V|^2. \tag{15}$$

From the data, we estimate $\delta\rho_s/\rho_s^0 \approx 1/4$ for $\delta n_3 \approx 10$ ppm. Assuming $n_V \approx 0.1\%$, we find $g \approx 25$.

CONCLUSION

Although we motivated our arguments with a wave function appropriate for considerations in the grand canonical ensemble, in our discussion of the effect of ^{3}He impurities on the superfluid stiffness, we assumed implicitly that changes in the average concentration of vacancies n_V with ^{3}He concentration are much less important than the decrease in the kinetic energy. We have no evidence for this but strictly speaking we cannot exclude the possibility that adding ^{3}He increases the equilibrium vacancy density. If this is the case, we need to augment our arguments by explicitly allowing n_V be dependent on

n_3. This would be an extension of our analysis that could lead to modifications of our results. Such an increase in n_V would be a simple source of the T_c increase with addition of ^{3}He. However, even the presence of patches of higher T_c could increase the bulk T_c by proximity effects. Similarly, the mass decoupling that is measured in the torsional oscillator would have higher onset temperature due to patches of higher T_c. We leave this issue until the experimental situation is clearer.

We presented a GL approach to the supersolid phase that contains two order parameters, density modulation $n_B(\mathbf{r})$ and superfluid $\psi_V(\mathbf{r})$. Both the solid and superfluid fields are of course made from the same ^{4}He atoms and realize a dual quantum mechanical behavior of these atoms. As others have argued, it seems reasonable to us that lattice defects, for example vacancies, are crucial for supersolid phenomena.

We find that disorder from ^{3}He atoms produces regions of depleted ^{4}He density, thus attracting vacancies. This attractive potential due to disorder can be though of as creating locally regions of higher T_c in the GL functional for ψ_V. At the same time the granularity in ψ_V produced by disorder leads to suppression of superfluid stiffness. Both of these effects are linear in concentration of ^{3}He atoms.

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[11] Furthermore, we believe that the activation energy for interstitials is larger than that for vacancies and the bandwidth of the former is likely to be smaller than that of the latter.
[12] This representation has been used for discussion of liquid ^{4}He in a number of works. See, for example, R.T. Whitlock and P.R. Zilsel, Phys. Rev. **131**, 2409 (1963); P.R. Zilsel, Phys. Rev. Lett. **15**, 476 (1965). A quantum lattice gas model also has a history in the literature. In particular, Liu and Fisher, J. Low Temp. Phys **10**, 655 (1973) discussed supersolid formation using a boson to spin mapping on a bipartite lattice. In their approach, the vacancy density does not appear as an explicit parameter hence ψ_V as a relevant order parameter does not enter. In contrast, our work focuses on the effects of disorder for nearly full ^{4}He lattice with a low vacancy density and an associated ψ_V order parameter.
[13] We are aware that simple nearest neighbor hopping may not be adequate to describe the multiparticle exchanges responsible for vacancy motion.
[14] The kinetic energy operator in the spin language is $(-t/2)\sum[\sigma_i \cdot \sigma_j - \sigma_i^z \sigma_j^z]$.
[15] According to Anderson, Brinkman and Huse [9], the T_c determined in the experiments is actually not a true transition temperature but some vortex dynamics temperature and that the true T_c cannot be determined directly from the torsional oscillator experiments. If this objection is correct then we cannot use existing data to estimate the λ coefficient. Nevertheless the prediction about T_c increase would be correct.